

Photoluminescence Spectroscopy of many-body Effects in Heavily Doped $\text{Al}_x\text{Ga}_{1-x}\text{As}$

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We present an experimental investigation of heavy doping-induced many-body effects such as band gap narrowing(BGN) and Fermi edge singularity(FES) in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ using photoluminescence(PL) spectroscopy. The band-to-band transition energy shifts to lower energies and FES-like feature in PL spectra grows with increasing electron concentration. We show that FES-like feature is a nonmonotonic function of temperature and excitation intensity. Our data lead us to suggest that a small concentration of nonequilibrated holes is required to enhance the FES-like feature in the PL spectra.

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There are two most important many-body effects related to heavy doping in III-V semiconductors, (i) the shift of conduction and valence band towards each other causing band gap narrowing(BGN) due to electron-electron interaction[1], and (ii) the many-body enhancement of the oscillator strength of the optical transition near Fermi level causing Fermi edge singularity(FES), due to the response of Fermi sea to a photogenerated hole[2]. The presence of the large concentration of free carriers can cause a significant reduction of unperturbed band gap in semiconductors due to many-body interaction between electrons and holes and carrier-impurity interaction[3]. Though a detailed experimental and theoretical analysis that rigorously combines the many-body effects due to exchange-correlation and statistical fluctuation is extremely complicated problem, one can investigate the role of many-body effects on BGN in alloy semiconductor by choosing the doping concentration carefully[3]. FES is one of the most widely investigated many-body effects, first theoretically predicted by Mahan[4]. This effect was first successfully understood in terms of threshold singularity in X-ray absorption and emission spectra in simple metals[5]. In case of semiconductors, it was first observed[6] in 2D electron gas in modulation doped $\text{InP}/\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum well and subsequently, in several other systems based on low dimensional semiconductor structures(LDSSs). It has been shown that the essential condition for observing the FES is the localization of the photogenerated holes. Typically the FES is identified as an enhancement of the oscillator strength of a transition close to Fermi level in PL spectra.

Although it appears that FES is the simplest nontrivial many-body effect and understood experimentally and theoretically, upon closer inspection it seems several experimental results such as (i) observation[7, 8] of FES in GaAs-based LDSSs without substantial hole localization, (ii) strong dependence[9, 10] of FES on carrier concentration and excitation intensity in GaAs- and InP-based LDSSs even in the presence of strong hole localization, and (iii) finally observation[11, 12] of FES at relatively higher temperature, are not understood within the existing theoretical framework. Moreover, LDSSs have

been used to observe FES, but there are two inherent shortcomings in these systems, (i) it is known that non-Coulombian intersubband scatterings can induce FES as well[13], so it is not known exactly the relative importance of the extrinsic intersubband scattering and intrinsic many-body electron-hole interaction on the formation of FES[14] and (ii) the exact role of hole localization and reduced dimension and their relative importance on the formation of FES is not completely understood in LDSSs.

In order to address these issues, we have investigated the formation of FES in higher dimensional systems, where the exact role of hole localization on FES may be understood unambiguously. If, the localization of photogenerated hole can be achieved, it should be possible to observe FES in degenerate bulk semiconductors[15]. A natural point to start, when studying heavy doping-induced many-body effect is BGN, which is probably the most well understood many-body effect and can be used to tune the range of electron concentration to observe the FES-like feature in PL spectra in heavily doped $\text{Al}_x\text{Ga}_{1-x}\text{As}$. We show first the BGN in moderately heavy doped n-type $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is governed by many-body effects. This has been validated by us in a previous study[3] using room temperature PL and reconfirmed in this work by low temperature PL study. This is then used to identify the FES-like feature in bulk $\text{Al}_x\text{Ga}_{1-x}\text{As}$. Emphasize is put on the anomalous temperature and excitation intensity dependence of FES. Finally a scenario based on the exchange splitting of FES i.e. many-body excitonic level has been invoked to model the nonmonotonic temperature dependence of FES.

Samples used in this investigation are high quality $\text{Al}_x\text{Ga}_{1-x}\text{As}$ epitaxial layers with doping level ranging from 3×10^{16} to $3 \times 10^{18} \text{cm}^{-3}$, grown on either semi-insulating and n-type GaAs substrate by metal organic chemical vapor epitaxy. Se is chosen as dopant for its high solubility, low compensation yielding highest doping level and minimum DX-center related problems[16]. The electron concentrations were determined by Hall measurements. The active $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers are separated from the GaAs substrate by an undoped spacer layer to avoid any 2D effect. Details about the sam-

ples are given in Ref.3. Room temperature and low temperature PL spectra of n-type $\text{Al}_x\text{Ga}_{1-x}\text{As}$ as a function of electron concentration for $x=0.22$ and $x=0.11$ are shown in Fig.1 and Fig.2, which show the evolution of low temperature PL spectra of $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$ as a function of electron concentration, respectively. In case of $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ ($\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$), the sample with lowest electron concentration of $3.5 \times 10^{16} \text{cm}^{-3}$ ($5.4 \times 10^{16} \text{cm}^{-3}$) in sample 1A(2A), the dominant peaks at 1.842eV(1.689eV) is due to bound exciton(BE) and the broad peak at 1.822eV(1.672eV) is due to free-to-bound(FEB)- and donor-acceptor(DAP)-related transitions. A high energy shoulder at 1.847eV(1.695eV) is due to either band-to-band(BB) or free exciton(FE). As the electron concentration is slightly increased to $4.5 \times 10^{16} \text{cm}^{-3}$ ($8.2 \times 10^{16} \text{cm}^{-3}$) in sample 1B(2B), almost similar PL spectra are observed in case of sample 1B(2B), but as the electron concentration increases to $1.3 \times 10^{17} \text{cm}^{-3}$ ($2.4 \times 10^{17} \text{cm}^{-3}$) in sample 1C(2C) BB peak appears in lieu of BE and FE peaks due to quenching of free excitons and screening of impurity potential. As the electron concentration is slightly increased to $3.1 \times 10^{17} \text{cm}^{-3}$ ($3.7 \times 10^{17} \text{cm}^{-3}$), almost similar PL spectra are observed in case of sample 1D(2D). Now, as electron concentration is further increased $8.3 \times 10^{17} \text{cm}^{-3}$ ($8.9 \times 10^{17} \text{cm}^{-3}$) in sample 1E(2E), an extra high energy peak at 1.858eV in addition to BB, FEB and DAP peaks is observed pronouncedly in sample $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and as weak a shoulder at about 1.724eV in sample $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$. Again, as the electron concentration is further increased to $9.6 \times 10^{17} \text{cm}^{-3}$ ($2.1 \times 10^{18} \text{cm}^{-3}$) slightly in sample 1F(2F), similar PL spectra as those in 1E(2E) is observed. Energetic position and saturation behavior have revealed the identification of these peaks[17]. Following are the general features of the PL spectra in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$.

(a) The transitions due to FE, BE, FEB and DAP are observed in samples with low electron concentration. As the electron concentration is increased by order of magnitude, FE and BE peaks are replaced with BB transition, but FEB and DAP peaks are observed in all samples.

(b) Typical BB transitions are observed in room temperature PL spectra(Fig.1 and Fig.2), which are characterized with Maxwell-Boltzman lineshape[3] in the high energy side and relatively sharp cut-off in the low energy side caused by $(E_G - E)^{1/2}$ dependence, where E_G is the band gap. It is clear that BB peaks shifts towards lower energy as the electron concentration increases.

(c) The redshift of BB peak is also observed at low temperature. Insets of Fig.1 and Fig.2 show the BGN as a function of electron concentration.

(d) The width of the all peaks, which are due to BB, FEB and DAP increases with electron concentration.

For determining the role of many-body effects due to exchange and correlation, it is desirable to restrict the

doping concentration such a way that typical BB lineshape can be observed in the PL spectra. It has been shown that the effective change in the unperturbed band gap E_G^0 due to many-body interactions can be given by[18], $E_G = E_G^0 - an^{\frac{1}{3}} - bn^{\frac{1}{4}}$, where n is electron concentration and first term is due to exchange interaction, which comes from the spatial exclusion of the like spin away from each other. The second term is due to correlation energy, which comes from the repulsion of the like charges, so that they do not move independently but in such a way as to avoid each other as far as possible. The values of the coefficients a and b , which are evaluated by fitting the experimental data(as shown in insets of Fig1. and Fig.2) with above expression are 1.6×10^{-8} and 8.2×10^{-8} in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and 8.2×10^{-9} and 2.1×10^{-8} in $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$, respectively. It is not possible to make any comments on the value of coefficients a and b either at 300K or at 1.8K, because neither detailed theoretical, nor experimental results are available on BGN in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. Hence BGN at high and as well as at low temperature governed by intrinsic many-body interaction i.e. exchange and correlation among the carriers.

Following are the important features of the high energy peaks observed in samples 1E and 1F in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and samples 2E and 2F in $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$

(a) The extra high energy peak at 1.858eV in case of sample $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ (1E and 1F) and a shoulder at 1.724eV in case of sample $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$ (2E and 2F) are not observed in previous PL studies on $\text{Al}_x\text{Ga}_{1-x}\text{As}$. The variation of the band-gap in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ due to alloy fluctuation determined by scanning the sample with bound exciton peak energy has been found to be small (~ 0.4 meV/cm) across the sample and this fluctuation corresponds the variation of x less than 0.001 per cm and rules out the possibility of energetic shift of BB or excitonic transition towards higher energy due to band gap variations.

(b) The energetic position of this new peak is almost 20meV above the band gap. We have found that the position of Fermi level in samples with electron concentration of $8.3 \times 10^{17} \text{cm}^{-3}$ to $9.6 \times 10^{17} \text{cm}^{-3}$ in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and $8.9 \times 10^{17} \text{cm}^{-3}$ to $2.1 \times 10^{18} \text{cm}^{-3}$ in $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$ is around 20meV at low temperature($< 10\text{K}$).

(c) The high energy peak is completely quenched in sample 1G with $2.8 \times 10^{18} \text{cm}^{-3}$ electron concentration, but BB, FEB and DAP peaks are present, as shown in Fig.1.

(d) Fig.3 shows the evolution of PL spectra of sample 1F($\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$), with excitation energy. At lowest intensity, the high energy peak is barely visible, but the spectral feature increases as excitation intensity increases. Further increase in excitation intensity smears out the peak.

(e) Fig.4 shows the temperature dependence of PL spectra for sample 1F($\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$). The temperature dependence of integral PL intensities of BB, FEB

and DAP peaks shows monotonic dependence, but the high energy peak shows completely opposite behavior. The integral intensity of this peak is observed to be a nonmonotonic function of temperature with a maximum at about 40K and this peak persists till higher temperature ~ 70 K. Similar nonmonotonic temperature dependence has also been observed(not shown) for high energy peak in $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$. Non-monotonic temperature dependence of FES-like peak results S-shaped shift behavior of BB peak. The deconvoluted BB peak, as shown in inset of Fig.4, shows usual temperature dependence.

All these findings indicate that the high energy peak is intrinsic in nature. We attribute the origin of this peak as due to many-body effect *i.e.*, FES, which gets further support from Fig.1 and Fig.2. This transition can be observed only in samples with narrow range of electron concentration of $8.3 \times 10^{17} \text{cm}^{-3}$ to $9.6 \times 10^{17} \text{cm}^{-3}$ in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ and $8.9 \times 10^{17} \text{cm}^{-3}$ to $2.1 \times 10^{18} \text{cm}^{-3}$ in $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$. This may be one of the reasons for not observing this FES-like feature in previous PL spectra of n-type $\text{Al}_x\text{Ga}_{1-x}\text{As}$. Disappearance of this peak in very heavily doped samples ($> 3 \times 10^{18} \text{cm}^{-3}$) may be due to two reasons, (i) it has been shown[19] that width of the luminescence spectra due to FES increases and the intensity decreases with increasing carrier density due to screening and movement of the electron layer from the localized hole and (ii) potential fluctuation due to the ionized donor can destroy the FES by smearing out the sharp Fermi surface when the doping is more than 10^{18}cm^{-3} . This is indirectly corroborated by our results on BGN at low and high temperature on the same samples. BGN is governed by mainly electron-electron interactions in those moderately doped samples ($n \leq 10^{18} \text{cm}^{-3}$) which show FES-like feature. We have observed that FES-like feature is completely quenched in samples with doping concentration more than 10^{18}cm^{-3} , which do not show typical BB lineshape. Here we would like to mention that weak appearance of FES in $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$ compared to that in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ may be due to the fact that alloy fluctuation in the dilute alloy is not enough to localize the photogenerated holes efficiently. Localization of photogenerated holes by alloy fluctuation plays an important role in the PL spectra of moderately heavy doped degenerate bulk semiconductor relaxing the \mathbf{k} -selection rule of photogenerated holes and provides the recombination channel with electrons occupying all \mathbf{k} -states upto Fermi-edge.

Fig.3 and Fig.4 show the temperature and excitation intensity dependences of the PL spectra of sample 1F. The observed nonmonotonic temperature and excitation intensity dependences of FES is not anticipated theoretically, because all previous theoretical investigations predict strong thermal quenching of FES due to smearing out of the Fermi surface. It is very clear that the nonmonotonic dependence of FES on temperature and excitation intensity implies that optimal tempera-

ture and excitation intensity are required for the observation of FES. Similar dependences of FES has been observed[11, 12] in pseudomorphically strained modulation doped $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{In}_y\text{Ga}_{1-y}\text{As}/\text{GaAs}$ heterostructures. Here, we present a scenario in which, nonmonotonic temperature dependence of PL intensity (I_{PL}) is modeled by two-level system. If we consider FES is more like localized many-body exciton, the nonmonotonic temperature dependence of I_{PL} can be described by exchange splitting of localized excitonic level[20] by an energy Δ between threefold degenerate triplet state and the higher lying singlet state, with radiative rate of R_T and R_S , respectively. At thermal equilibrium the total radiative decay R_R can be given by $R_R = \frac{3R_T + R_S \exp(-\Delta/k_B T)}{3 + \exp(-\Delta/k_B T)}$. At low temperature only the lower lying triplet state is occupied and the radiative decay rate is small, because the optical transition from a pure triplet state is forbidden, but, spin-orbit interaction mixes triplet and singlet states and allows optical transition with very low quantum yield. As the temperature increases, higher lying singlet state is occupied and quantum yield of the optical transition or the I_{PL} of FES-like peak increases and attains a maximum, but, further increase in temperature leads to thermal quenching of the bound states resulting the decrease of I_{PL} resulting nonmonotonic temperature dependence of FES. Inset of Fig.4 shows the fit to the experimental data with our phenomenological model. We have obtained a value of 6meV for Δ . We have found that the Δ for exciton is close to 0.5meV in $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$, but the localization of exciton can enhance this value drastically, for example Δ in GaAs is enhanced by more than two orders of magnitude for excitons confined in a quantum dot[21]. Further experiments are required to corroborate this scenario.

In conclusion, we have observed many-body effects like BGN and FES-like features in moderately heavy doped $\text{Al}_x\text{Ga}_{1-x}\text{As}$ by PL spectroscopy. FES arises due to multiple scattering of electrons near Fermi-edge by the photogenerated holes localized by alloy fluctuation in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. A two-level-based model has been proposed to can explain the nonmonotonic temperature and excitation intensity dependence of FES related transition in PL spectra. The present results indicate two directions for future research. More experimental and theoretical are required for further ascertainment of (i) the nonmonotonic temperature and excitation intensity dependence of FES and (ii) the effect of hole localization on many-body effects leading to the formation of FES.

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Figure Captions

Fig.1. Evolution of PL spectra of $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ as a function of electron concentration. Solid lines(for samples 1A, 1B, 1C, 1D, 1E, 1F, 1G) represent PL spectra at $T=1.8\text{K}$ and dashed lines(for samples 1B and 1D) represent PL spectra at $T=300\text{K}$. Triangles and inverted triangles indicate the spectral position of band-to-band

transition at $T=300\text{K}$ and 1.8K , respectively. Inverted arrows(\Downarrow) indicate the spectral position of the FES-like feature in PL spectra. Inset shows the reduction of band gap as a function of logarithm of electron concentration.

Fig.2. Evolution of PL spectra of $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}$ as a function of electron concentration. Solid lines(for samples 2A, 2B, 2C, 2D, 2E, 2F) represent PL spectra at $T=1.8\text{K}$ and dashed lines(for samples 2C and 2E) represent PL spectra at $T=300\text{K}$. Triangles and inverted triangles indicate the spectral position of band-to-band transition at $T=300\text{K}$ and 1.8K , respectively. Inverted arrows(\Downarrow) indicate the spectral position of the FES-like feature in PL spectra. Inset shows the reduction of band gap as a function of logarithm of electron concentration.

Fig.3. Excitation-intensity dependence of photoluminescence spectra of $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ with $9.6 \times 10^{17} \text{cm}^{-3}$ (sample 1F) electron concentration at 1.8K . Arrows(\Downarrow) indicate the spectral position of the FES-like feature in PL spectra. Inset shows the PL intensity of FES-like feature as a function of logarithm of excitation intensity(I). Solid line is guide for the eyes.

Fig.4. Temperature dependence PL spectra of $\text{Al}_{0.22}\text{Ga}_{0.78}\text{As}$ with $9.6 \times 10^{17} \text{cm}^{-3}$ (sample 1F) electron concentration. The emission peak due to band-to-band transition shows an anomalous S-shaped shift with increasing temperature(shown as solid circles) due to the nonmonotonic temperature dependence of PL intensity of high energy FES-like peak, shown in upper inset. Lower inset shows the deconvoluted BB peak, which shows usual monotonic redshift with increasing temperature.

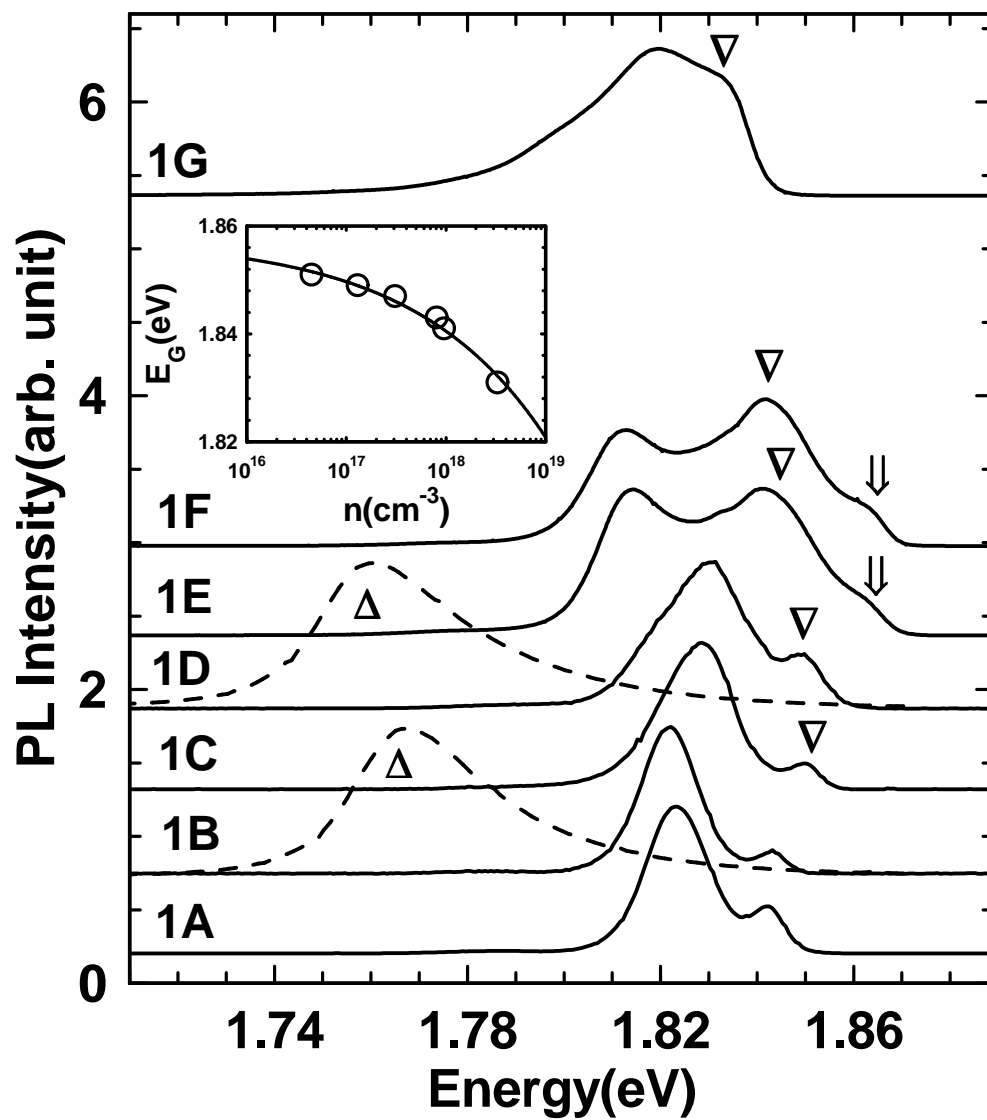


Figure 1

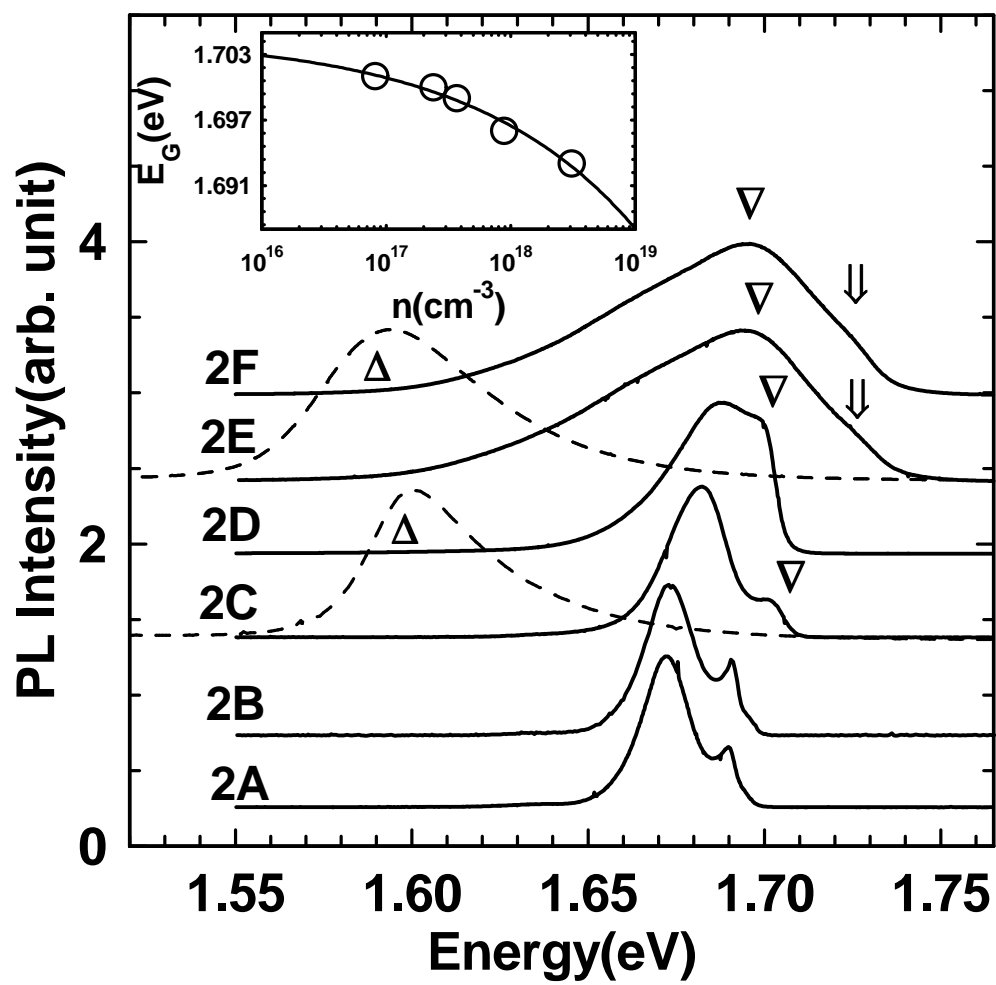


Figure 2

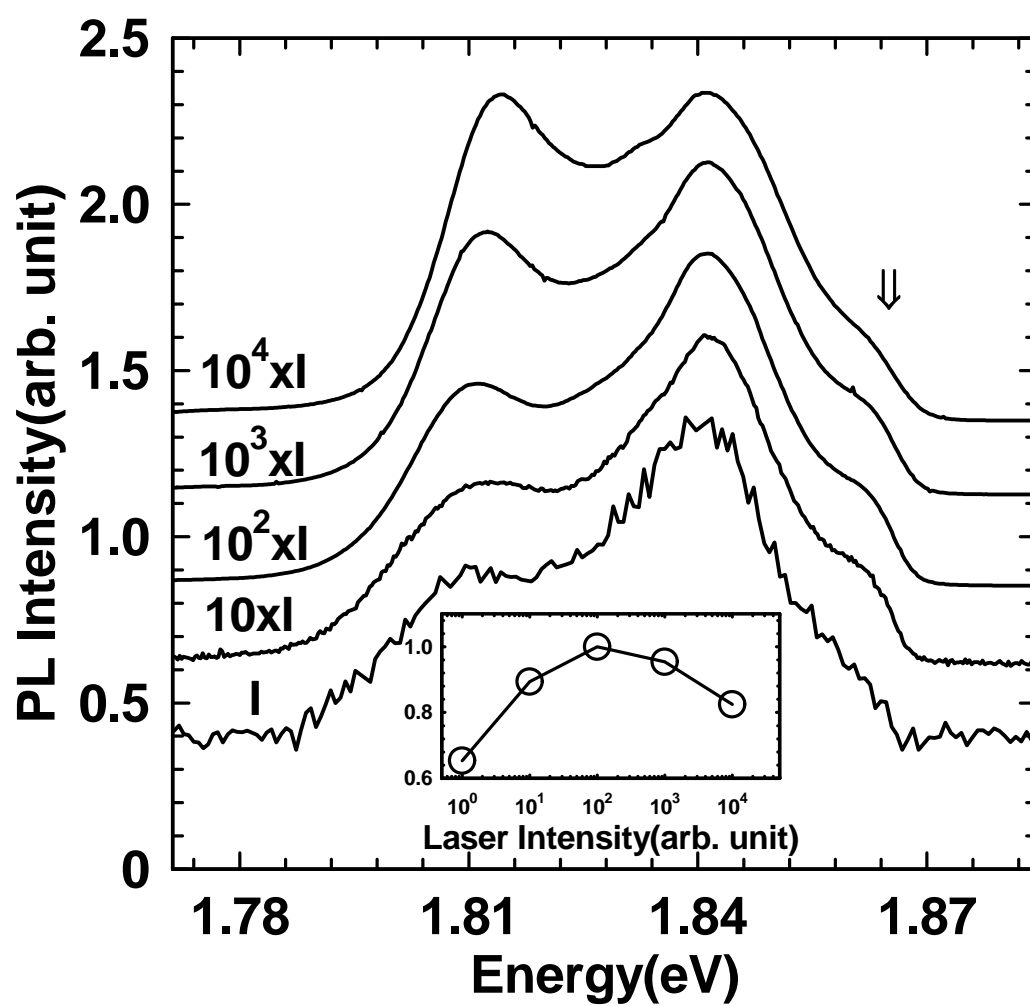


Figure 3

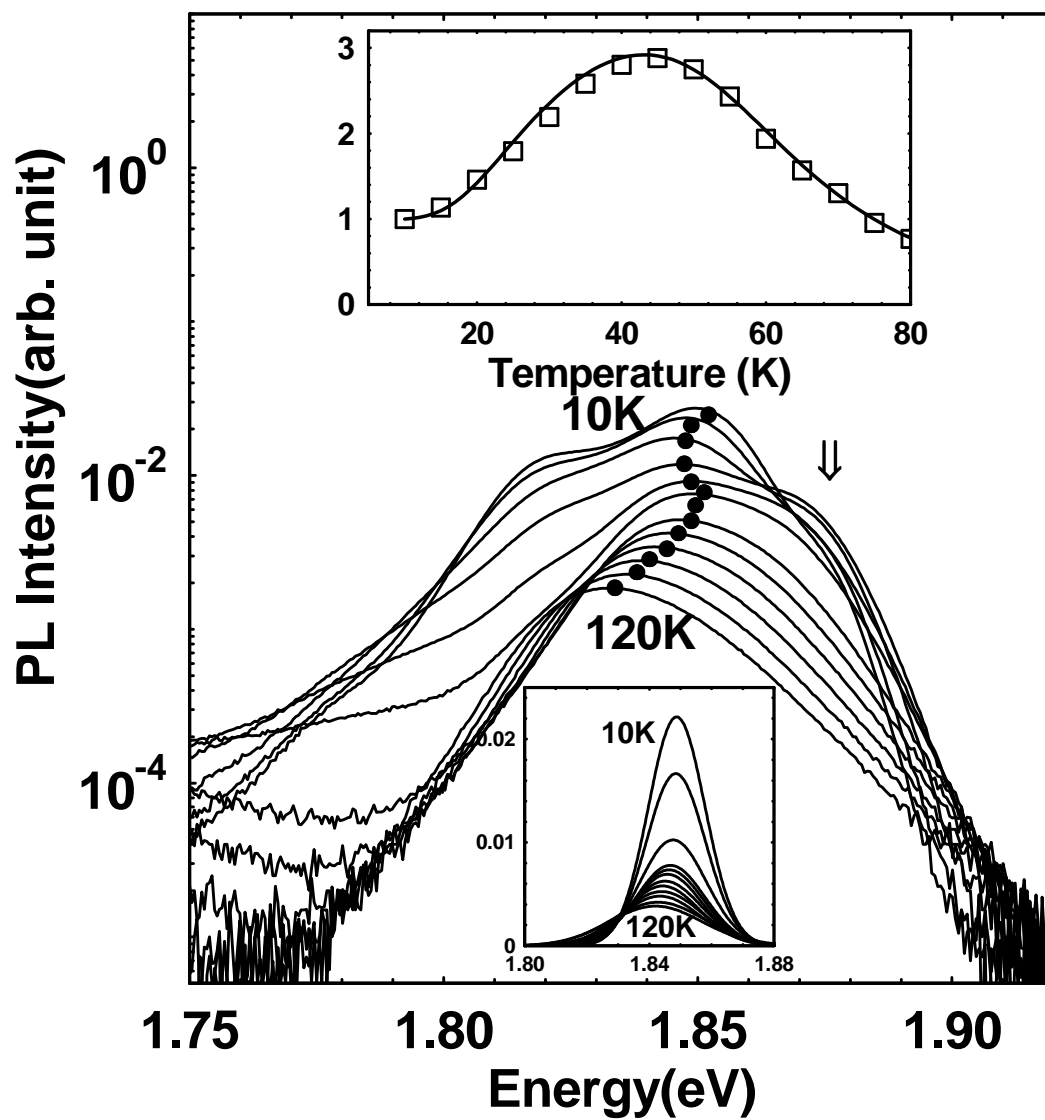


Figure 4